

Collisions of cold magnesium atoms in a weak laser field

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We use quantum scattering methods to calculate the light-induced collisional loss of laser-cooled and trapped magnesium atoms for detunings up to 30 atomic linewidths to the red of the 1S_0 - 1P_1 cooling transition. Magnesium has no hyperfine structure to complicate the theoretical studies. We evaluate both the radiative and nonradiative mechanisms of trap loss. The radiative escape mechanism via allowed $^1\Sigma_u$ excitation is dominant for more than about one atomic linewidth detuning. Molecular vibrational structure due to photoassociative transitions to bound states begins to appear beyond about ten linewidths detuning.

34.50.Rk, 34.10.+x, 32.80.Pj

Light-induced collisions between cold, neutral alkali atoms have been widely studied experimentally and theoretically in magneto-optical traps [1–3]. Such collisions cause loss of atoms from the trap due to radiative or nonradiative molecular processes after laser excitation of molecular states of the atom pair at large internuclear separation R . These loss processes for alkali atoms are still not well-understood when the laser detuning Δ is only a few natural atomic linewidths Γ_{at} to the red of atomic resonance, primarily because of complications due to the molecular hyperfine structure in the alkali species [3,4]. Molecular vibrational and rotational structure in the spectrum of loss versus Δ is unresolved due to broadening by fast spontaneous decay and the high density of molecular states. On the other hand, high resolution photoassociation spectra for trap loss at much larger detunings are quite well-understood quantitatively, even for alkali species [3,5].

The study of trap loss collisions in alkaline earth species offers a significant opportunity to improve our understanding of collisional loss processes in the small detuning regime. The major isotopes of Be, Mg, Ca, Sr, and Ba have no nuclear spin, and thus lack hyperfine structure. Alkaline earth atoms can also be cooled and trapped (e.g. Mg [6], Ca [7], Sr [7,8]). They offer an excellent and previously rather unexplored testing ground for cold collision theories. A recent study of Sr has taken a first step in this direction [9]. One intriguing aspect of the alkaline earth collisions is the possibility to observe the effect of relativistic retardation corrections on the trap loss dynamics by exciting a molecular state with a dipole-forbidden transition at small R which becomes

allowed at large R . This predicted effect for the 2_u state of alkali species [10] is masked by other states, but may be prominent for the $^1\Pi_g$ state of alkaline earth species.

In this paper we calculate collisional loss rate coefficients of Mg atoms in a magneto-optical trap, induced by a weak laser field detuned up to $30\Gamma_{\text{at}}$ to the red of atomic resonance ($\Gamma_{\text{at}} = 2\pi \cdot 80$ MHz). This spans the range from small detuning, where the spectrum is unstructured, to the far-detuning range, where resolved photoassociative features due to specific molecular vibrations begin to appear. For ^{24}Mg we assume that the cooling transition is 1S_0 - 1P_1 , ($\lambda = 285.2$ nm). Fig. 1 shows the short-range part of the Mg_2 potentials [11]. A laser with $\Delta \sim 1$ - $30\Gamma_{\text{at}}$ excites molecular states with attractive potentials at some Condon distance $R_C \gtrsim 250 a_0$ ($a_0 = 0.0529177$ nm). In ^{24}Mg these states can be either the $^1\Sigma_u^+$ state or the $^1\Pi_g$ state. Spin-orbit coupling H_{SO} mixes $^1\Pi_g$ with the $^3\Sigma_g$ state at a curve crossing around $R \sim 7 a_0$ and mixes $^1\Sigma_u^+$ with $^3\Pi_u$ around $R \sim 5 a_0$. After a change of molecular state the quasimolecular atom pair can separate to a 1S_0 atom and a 3P atom (3P_0 , 3P_1 or 3P_2), with a shared kinetic energy increase equal to about 0.06 atomic units ($\Delta E/k_B \sim 19\,000$ K). These hot atoms can not be recaptured by the trapping and cooling lasers and they are thus lost from the trap. In alkalis this process arises from collisional transfer between two fine structure states of the same electronic configuration, and is called the fine structure changing mechanism (FS). Here we call it the state changing mechanism (SC) between different electronic states.

After excitation at R_C the quasimolecule obtains kinetic energy via internal acceleration by the attractive potentials. If it decays back to the ground state, it preserves this gain in kinetic energy, thereby increasing the kinetic energy of the asymptotic collision products (two 1S_0 atoms). If the energy gain exceeds the trap depth, the atoms can escape from the trap. This is the radiative escape mechanism (RE). Spontaneous emission has a crucial role in both the SC and RE loss mechanisms, and has to be included in the theoretical treatment.

The structure of the quasimolecule at large R is determined by the dipole-dipole interaction, for which we use the long-range retarded potentials from Ref. [12]. These join with the short-range wells in Fig. (1) to give potentials that support a number of bound vibrational states of the quasimolecule. The molecular linewidths with relativistic retardation corrections for the two relevant ex-

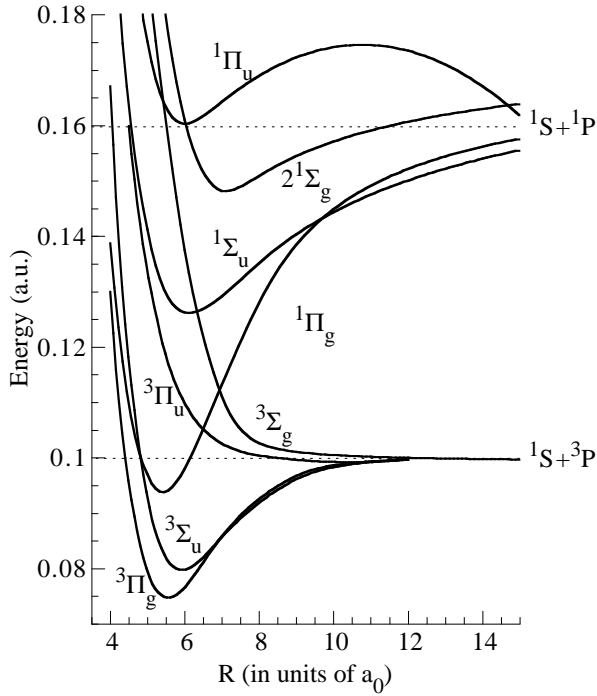


FIG. 1. The molecular states of Mg_2 in atomic units (1 a.u. = $4.3597482 \times 10^{-18}$ J) corresponding to the asymptotic atomic states $^1\text{S}_0 + ^1\text{P}_1$ and $^1\text{S}_0 + ^3\text{P}_{0,1,2}$ [11]; the zero of energy is at the ground state $^1\text{S}_0 + ^1\text{S}_0$ asymptote. There are four states related to each asymptote, of which two are attractive and two are repulsive at large R , where the system is expected to be resonant with the laser field. Each set of two states has one Π state and one Σ state.

cited states with attractive potentials are [12]

$$\Gamma_{\Sigma_u}(R) = \Gamma_{\text{at}} \left\{ 1 - \frac{3}{u^3} [u \cos(u) - \sin(u)] \right\}, \quad (1)$$

$$\Gamma_{\Pi_g}(R) = \Gamma_{\text{at}} \left\{ 1 - \frac{3}{2u^3} [u \cos(u) - (1 - u^2) \sin(u)] \right\}, \quad (2)$$

where $u = R/\lambda$, and $\lambda = \lambda/2\pi = 857.7 a_0$. For small u we get $\Gamma_{\Sigma_u} \simeq \Gamma_{\text{at}}(2 - u^2/10)$ and $\Gamma_{\Pi_g} \simeq \Gamma_{\text{at}}u^2/5$. Note that the optical coupling between the quasimolecule and the laser field is proportional to $\sqrt{\Gamma_A}$ ($A = \Sigma_u, \Pi_g$). As detuning Δ increases the probability to excite the $^1\Pi_g$ state is reduced. On the other hand, the survival of the quasimolecule on the $^1\Pi_g$ state is much better than for the $^1\Sigma_u$ state. We expect that RE loss comes mainly from the $^1\Sigma_u$ state, and that qualitatively the $^1\Pi_g$ state and the $^1\Sigma_u$ state contributions to SC loss behave differently as a function of Δ . The important question is the relative magnitudes of the various loss mechanisms.

The standard small-detuning studies of trap loss ignore the vibrational structure of the trap loss, and consider the problem as a fully dynamical one, involving only the electronic potentials [2]. This is based on the expectation that at small detunings the vibrational structure can not be resolved. In the time-dependent picture this simply means that after the excitation at large R the quasi-

molecule decays well before it completes one or more vibrations. However, here the retardation effect opens the $^1\Pi_g$ state for excitation but closes it quickly for decay, so we might expect to see vibrational structure in trap loss even at small detunings if the $^1\Pi_g$ state SC contribution dominates over the $^1\Sigma_u$ state SC and RE contributions.

The ground state potential of Mg_2 is essentially flat in the long-range region of excitation. Since a number of total angular momentum J values contribute to trap loss in our detuning range, we assume that effects of the unknown phase shifts in the ground state wavefunctions due to the short-range potential is removed by summing over J and thermal averaging. Thus, we represent the ground state potential by a Lennard-Jones 6-12 potential with a well depth of 0.0028 atomic units and an inner turning point of $6.23 a_0$ [11]. Since the excited *ab initio* potentials do not permit a calculation of vibrational levels to spectroscopic accuracy, the specific forms of the short-range excited state potentials are not important for our purposes of modeling the qualitative structure and magnitude of the collisional loss. We fit Lennard-Jones 3-6 potentials to the *ab initio* potentials of Ref. [11], keeping the long-range form fixed to its known value. Modeling of experimental data should ultimately permit calibration of the unknown phases associated with the uncertain inner parts of the potentials.

A complete study must include the three-dimensional aspects of molecular rotation. Since all collision directions are possible relative to laser polarization, the effect of axis rotation is to introduce the usual molecular branch structure of the possible transitions. The angular momentum J'' in the ground state can only be that of axis rotation, or partial wave $\ell'' = J''$. In the excited state $^1\Sigma$, $^1\Pi$ molecular basis, ℓ is not a good quantum number but total angular momentum J' is. The possible transition branches are P, Q, and R, for which $J' = J'' - 1$, J'' , and $J'' + 1$ respectively. The quasimolecule ground state can couple to the $^1\Sigma_u$ state only by P and R branches, but to the $^1\Pi_g$ state by P, Q and R branches. The radiative coupling terms for the two electronic states for each branch and partial wave ℓ'' are shown in Table I.

State A	Branch B	$\ell'' = 0$	$\ell'' \neq 0$
Σ	P	0	$\sqrt{\ell''/3}$
Σ	R	$\sqrt{2/3}$	$\sqrt{(\ell'' + 1)/3}$
Π	P	0	$\sqrt{(\ell'' - 1)/3}$
Π	Q	0	$\sqrt{(2\ell'' + 1)/3}$
Π	R	$2/\sqrt{3}$	$\sqrt{(\ell'' + 2)/3}$

TABLE I. The radiative coupling weight factors $\alpha_{A,B,\ell''}$ for each electronic state, ground state partial wave ℓ'' and excitation branch, obtained by summing over the degenerate projection states. The full radiative coupling in atomic units is $V_{A,B,\ell''} = 1.16 \times 10^{-4} \alpha_{A,B,\ell''} \sqrt{I \text{ (W/cm}^2\text{)}} \sqrt{\Gamma_A(R) \text{ (a.u.)}}$.

Since the laser field is assumed to be weak, reexcitation of any decayed quasimolecular population can be ignored and loss rates can be calculated by the complex potential method [13,14]. Additionally, each ground state partial wave only couples to the rotational states of the $^1\Sigma_u$ or $^1\Pi_g$ state, but these states do not couple further to other ground state partial waves. Thus, the weak field approximation permits us to truncate the problem into a set of independent three-state problems for each initial partial wave with one ground state g , one excited state e , and one probe state p . The total thermally averaged loss rate coefficient via state e is

$$K(\Delta, T) = \frac{k_B T}{h Q_T} \int_0^\infty \frac{d\varepsilon}{k_B T} \exp\left(-\frac{\varepsilon}{k_B T}\right) \sum_{l''_{\text{even}}} (2l'' + 1) \times \sum_{B=P,Q,R} |S_{gp}(\varepsilon, \Delta, l'', B)|^2. \quad (3)$$

where ε is the relative asymptotic initial kinetic energy, $Q_T = (2\pi\mu k_B T/h^2)^{3/2}$ is the translational partition function (μ is the reduced mass of the two colliding atoms), and $|S_{gp}|^2$ represents the probability of a transition to a probe channel p which simulates the effect of the SC or RE exit channels [14]. Identical particle exchange symmetry ensures that only even partial waves exist for the ground state. The ground state centrifugal potential provides a natural cutoff ℓ_{max} to the sum over l''_{even} in Eq. (3) where $\hbar^2 \ell_{max}(\ell_{max} + 1)/2\mu R_t^2 > \varepsilon$ at the classical turning point R_t for collision energy ε .

We calculate $S_{gp}(\varepsilon, \Delta, l, B)$ using a complex potential [13,14], i.e., spontaneous decay is taken into account by adding an imaginary decay term $-i\hbar\Gamma_A(R)/2$ ($A = \Sigma_u, \Pi_g$) to the excited state potentials. We solve the corresponding time-independent Schrödinger equation with the appropriate boundary conditions and obtain the S -matrix elements.

For the SC loss studies the probe state represents the actual loss channel due to the curve crossings mentioned above which cause transitions to the molecular states which separate to $^1S_0 + ^3P$ atoms. The short-range molecular spin-orbit couplings are approximated using Table A1 of Ref. [15]. The $^1\Sigma_u - ^3\Pi_u$ and $^1\Pi_g - ^3\Sigma_g$ matrix elements are $\lambda/\sqrt{2}$ and $\lambda/2$ respectively, where $\lambda = 1.84 \times 10^{-4}$ atomic units is 2/3 of the atomic $^3P_2 - ^3P_0$ splitting.

For the RE studies we assume that the trap depth is 0.5 K, and set the probe state to cross the excited quasimolecular state at the point R_e where a kinetic energy increase of 1 K for the atom pair has been gained after excitation. Equation (5) of Ref. [14] shows that $|S_{gp}|^2$ can be factored into two terms: $|S_{gp}|^2 = J_Q P_{\text{decay}}$, where J_Q is the probability of being excited from g and reaching R_e on e (J_Q accounts for the possibility of multiple vibrations). J_Q is calculated from $|S_{gp}|^2$ as explained in Ref. [14], and P_{decay} , the total probability of radiative escape during a vibration in the region $R < R_e$, is calculated using a classical trajectory:

$$P_{\text{decay}} = 1 - \exp(-a), \quad a = 2 \int_{R_e}^0 dR \frac{\Gamma_{\Sigma_u}(R)}{v(R)}; \quad (4)$$

$\Gamma_{\Sigma_u}(R)$ is given by Eq. (2), and $v(R)$ is the classical speed related to the $^1\Sigma_u$ state potential.

Fig. 2 shows calculated rate coefficients K for SC loss for both states at laser intensity $I = 1$ mW/cm². K scales linearly with I in the weak field limit. When Δ is small, the magnitude of K for the $^1\Sigma_u$ state is small and vibrational state structure is clearly suppressed even before thermal averaging; some structure begins to appear beyond $\Delta \approx 10\Gamma_{\text{at}}$. However, for the $^1\Pi_g$ state strongly modulated vibrational structure persists over the whole range of detuning with comparable peak magnitudes. The $^1\Pi_g$ vibrational structure is broadened but not eliminated by the sum over rotational state structure and by thermal averaging. The specific vibrational peak locations versus Δ for both states depend on our particular choice for the excited state potentials and may be expected to shift if the correct potentials were used. However, the magnitudes and qualitative nature of the oscillations will be independent of this choice. For very small detunings the $^1\Pi_g$ state dominates SC loss, due to much more favorable survival after excitation. For $\Delta \gtrsim 10\Gamma_{\text{at}}$ $^1\Sigma_u$ and $^1\Pi_g$ contributions become roughly equal. The survival probability relative to spontaneous decay at long range approaches unity for the $^1\Sigma_u$ state when $\Delta \gtrsim 10\Gamma_{\text{at}}$. Note that the magnitude of the rate coefficients for both $^1\Sigma_u$ and $^1\Pi_g$ states is rather small, on the order of 10^{-14} cm³/s. This is because of the relatively small spin-orbit matrix elements at the short-range crossings.

The RE loss for the $^1\Sigma_u$ state, shown in Fig. 3, qualitatively follows that of the corresponding $^1\Sigma_u$ state SC loss (see the inset in Fig. 3), but is about 200 times stronger. Because of this inherently larger magnitude for RE loss, trap loss for Mg is predominantly due to the $^1\Pi_g$ state only for very small detunings on the order of Γ_{at} . Radiative escape via $^1\Sigma_u$ excitation is dominant at all larger detunings and masks any vibrational structure due to $^1\Pi_g$ SC loss. Vibrationally resolved (but not rotationally resolved) photoassociative structure should begin to appear due to $^1\Sigma_u$ excitation for detunings larger than around 1 GHz, with a rate coefficient on the order of 10^{-12} cm³/s for $I = 1$ mW/cm².

Our calculations for Mg shed some light on Sr collisions. Since the spin-orbit coupling strength λ is a factor of 9.6 larger for Sr than for Mg, the SC loss will be much stronger in Sr than we find here for Mg, and thus will compete with RE as a loss mechanism. If we simply scale up our peak magnitude of $^1\Pi_g$ loss at $\Delta = 4\Gamma_{\text{at}}$ linearly in intensity I and by the increased λ^2 factor (differences in mass also need to be taken into account), the resulting rate coefficient has the same order of magnitude as that which was recently measured [9]. This suggests that Sr trap loss at small detuning, unlike that for Mg, has a prominent contribution from SC by the $^1\Pi_g$ state, consistent with the understanding of Ref. [9]. Since it is likely

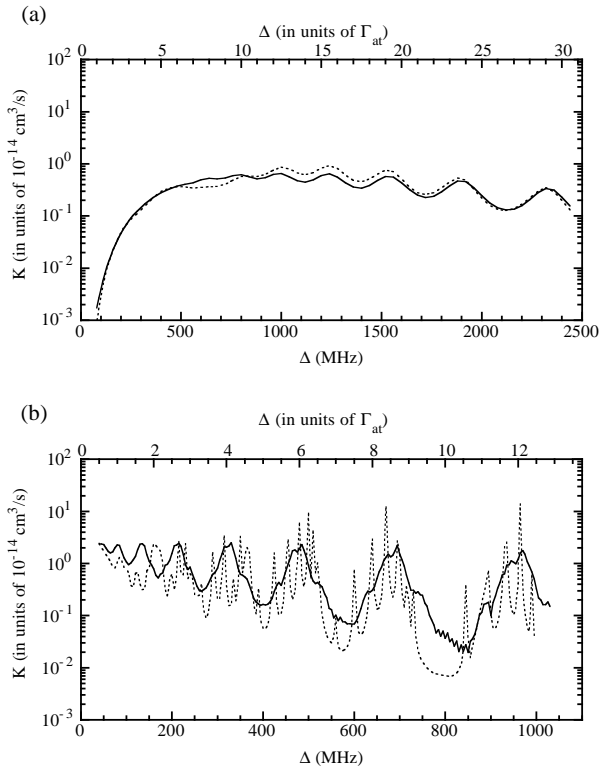


FIG. 2. The SC loss rate coefficient K_{SC} as a function of laser detuning Δ for Mg at laser intensity $I = 1 \text{ mW/cm}^2$ and $T = 2 \text{ mK}$ (the Doppler temperature for the chosen Mg cooling transition is $T_{\text{Doppler}} = 1.9 \text{ mK}$). The dotted lines show the sum over branches and partial waves for $\varepsilon = k_B T$, and the solid lines are the energy averaged result. a) $^1\Sigma_u$ state, b) $^1\Pi_g$ state.

that Sr will have a modulated photoassociation spectrum at small Δ analogous to Fig. 2b, measuring trap loss versus Δ would be a very useful experiment to try for the Sr system. Such modulations might also be observed in both Sr and Mg by detecting $^3P_{0,1,2}$ product atoms, as in similar alkali experiments [4,16].

Exploring the alkaline earth systems offers the prospects for some very fruitful science. There is clearly a need for better *ab initio* or semiempirical potentials and long-range dispersion coefficients. Photoassociation spectroscopy over a wide range of detuning values should help characterize ground and excited state potentials and ground state scattering lengths [3]. The recent cooling of Sr below $1 \mu\text{K}$ using the 1S_0 - 3P_1 transition [8] raises the possibility of achieving Bose-Einstein condensation with an alkaline earth species. Comparing experimental trap loss for alkaline earth isotopes with and without hyperfine structure might also shed some light on the role of hyperfine structure in alkali trap loss.

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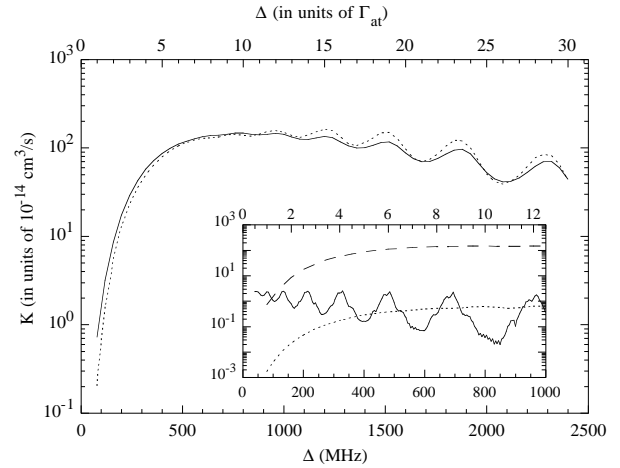


FIG. 3. The RE loss rate coefficient K_{RE} for the $^1\Sigma_u$ state as a function of the red detuning Δ for Mg at laser intensity $I = 1 \text{ mW/cm}^2$ and $T = 2 \text{ mK}$. The trap depth is set to 0.5 K . The inset shows the temperature averaged results for the loss rate coefficients K for all three loss processes (short dash, solid, and long dash lines are from Figs. 2a, 2b, and 3 respectively).

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